

Absolute Cosmic Ray Ionization Measurements in a 900-Liter Chamber

GRANVIL C. KYKER, JR.

*Division of Physics, Electrical Engineering and Computer Sciences, Rose-Hulman Institute of Technology
Terre Haute, Indiana 47803*

ABRAHAM R. LIBOFF

Department of Physics, Oakland University, Rochester, Michigan 48063

We report a new measurement for the cosmic ray sea level ionization, 2.15 ± 0.05 in ion pairs/s cm², in agreement with the previous determination by Shamos and Liboff (1966) (2.18 ± 0.05 ion pairs/s cm²) and the calculation by O'Brien (1970) for solar minimum (2.22 ion pairs/s cm²). In contrast with the 1966 measurement (which used CCl₂F₂), the present work was done in an air-filled 906-l aluminum chamber, with a wall thickness of ~ 0.3 g/cm². Measurements were conducted over Lake St. Clair and Lake Huron during 1973 and 1974. Corrections for alpha and beta wall contamination were made in two independent ways, by an analysis of the ion current fluctuations and by operating the chamber in the low-background Morton Salt Company Mine, located 600 m below ground in Painesville, Ohio.

INTRODUCTION

It has been difficult [Lowder and Beck, 1966; George, 1970; Raft *et al.*, 1970] to obtain a consensus on how best to measure the cosmic ray ionization intensity in the lower atmosphere. Theoretical calculations by O'Brien [1970] and O'Brien and McLaughlin [1972] based upon a high-energy transport code tend to support, at a given altitude, the lower values of the range of reported measurements. Experimental corroboration of O'Brien's altitude variation, at least for the lower altitudes, has recently been shown by Liboff [1975]. To explain in part the observed discrepancies, Carmichael [1971] has suggested that there have been consistent calibration errors in measurements at the California Institute of Technology, dating as far back as 1928 and amounting at times to as much as 32%.

In the following, we address ourselves to the directly related problem of determining the cosmic ray ionization intensity at sea level. Lying at the end of the altitude variation, it is highly important for normalization purposes. However, unavoidable experimental difficulties exist near sea level. Whereas at high altitudes the cosmic ray intensity increases rapidly, near the earth the available signal is so small that the terrestrial background radiation and any detector contamination become much more troublesome, and accurate measurements are difficult.

Some idea of this difficulty may be gained from examining Table 1, which lists most, if not all, of the reported thin-walled ionization chamber measurements on the sea level cosmic ray ionization intensity at high latitudes. Consider the four most recent values, at the top of Table 1. These determinations range from 2.1 to 2.6 ion pairs/sec cm² of STP air. In three cases thin plastic walls were used, and in the fourth an attenuation correction was made for the steel wall; it does not seem that wall effects—scattering or attenuation—can explain away the large discrepancies. However, it is generally agreed that radioactive impurities in the chamber walls can have a strong effect on this type of measurement. The chambers in question had quite different propensities for contamination. The Gustafson *et al.* [1964] chamber had conducting plastic walls, and Lowder and Beck [1966] chamber steel walls; Shamos and Liboff [1966] used a thin carbon coating on plastic as an outer electrode, and George [1970] used aluminized Mylar. The gas

fillings were also diverse: tissue equivalent gas at atmospheric pressure, argon at high pressure, Freon 12, and air at atmospheric pressure, respectively. Two of these devices were aimed specifically at eliminating or minimizing the effect of wall radioactivity. Using a high gas pressure reduces the relative contribution of ionization from wall contaminants; the use of a highly electronegative gas, together with a proper choice of collecting voltage, selectively enhances columnar recombination of the dense tracks of wall alpha particles [Shamos and Liboff, 1968]. These techniques, while dealing effectively with alpha contamination, do little to reduce the unwanted ionization due to betas from the walls. It will be seen below that the effect of beta activity in the chamber walls cannot be ignored.

Note that the four values at the bottom of Table 1 represent independent checks of earlier listed measurements, using either the identical original chamber or a close facsimile. The results of Yeates *et al.* [1972] are especially interesting, since they confirm the discrepancy between the results of Shamos and Liboff [1966] and Gustafson *et al.* [1964].

It is unlikely that the differences in Table 1 are due to any external noninstrumental physical phenomenon. The geomagnetic latitude, for example, is roughly the same for the various observers and in most cases is well into the cosmic ray polar plateau. The variation is not due to solar cycle modulation. The recent measurements were carried out during solar cycles 19 and 20; the latter began in mid-1964 and maximized in late 1968. Lowder *et al.* [1971] reported measuring 5% less than in the 1965 experiments, whereas George's data, yielding the largest of the recent results, were taken in 1968. Furthermore, O'Brien [1970] estimates the total variation in sea level cosmic ray ionization, during a solar cycle, to be of the order of 10%. Therefore after solar modulation effects are accounted for, there remains a 30% variation in the measurements reported.

There is much to be said for the approach taken by George. Because everyone's results are reported normalized to standard air, a direct measurement in air has the virtue of avoiding the question of the relative ionization to that in air of gases such as freon or argon. Thus the freon-air ratio used by Shamos and Liboff has been criticized by George; further, a wide range of values for the argon-air ratio, from 0.62 to 0.72, has been reported. The walls of George's chamber were sufficiently thin that wall effects of any kind must be negligible, and its active volume of 552 l was large enough to reduce

TABLE 1. Measurements of the Cosmic Ray Ionization at Sea Level Using Thin-Walled Ionization Chambers

Observers	Value* ion pairs/ s cm ²	Period of Measurement	Ionization Chamber Details				Comments
			Gas Filling	Active Volume, l	Internal Pressure, atm	Wall Thickness, g/cm ²	Wall Material
<i>George</i> [1970]	2.60	spring 1968	air	552	1+	0.013	aluminized Mylar, epoxy, aluminum
<i>Lowder and Beck</i> [1966]	2.1	fall 1964 to summer 1965	argon	5.58	54	2.7	steel
<i>Shamos and Liboff</i> [1966]	2.18	summer 1959 to fall 1964	freon-12	16.02–129.4	1+	0.5–1.2	graphite and plexiglas
<i>Gustafson et al.</i> [1964]	2.5	summer 1963	tissue equivalent gas mixture†	16.5	1	0.64	tissue equivalent conduct- ing plastic
<i>Solon et al.</i> [1960]	2.2	spring 1958	air	20	1	1+	graphite polyethylene aluminum
<i>Hess and Vancour</i> [1950]	1.91	summer 1949	air	1.6–43.7	1	2.0	brass
<i>Millikan and Neher</i> [1936], <i>Neher</i> [1952]	3.06	summer 1934	argon	1.622	30	2.4	steel
<i>Millikan and Cameron</i> [1931], <i>Millikan</i> [1932]	2.77	summer 1928	air	1.622	26.8	2.4	steel
<i>Yeates et al.</i> [1972]	2.57	summer 1969	tissue equivalent gas mixture†	16.5	1	0.64	tissue equivalent conduct- ing plastic
<i>Yeates et al.</i> [1972]	2.06	summer 1969	freon-12	16.02	1+	0.5	graphite and plexiglas
<i>Ohlsen</i> [1969]	1.9	1965–1966	air	23	1	1+	graphite polyethylene aluminum
<i>Herbst and Hübner</i> [1961], <i>Herbst</i> [1964]	2.1	summer 1960	air	25	1	1+	graphite polyethylene aluminum

*In STP air.
†By volume: 41.11% neon, 39.59% ethylene, 16.17% ethane, 3.13% nitrogen.

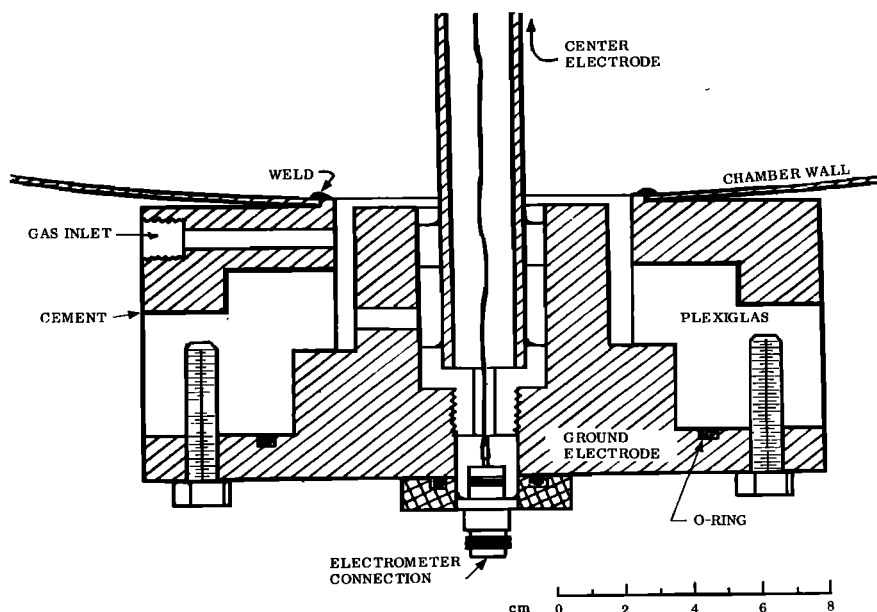


Fig. 1. Terminal detail of 906-I ionization chamber.

substantially whatever wall alpha current may have been present. (Recall that since alpha particles are stopped in a few centimeters of STP air, their contribution to the chamber current varies as the ratio of surface area to volume, i.e., inversely as the radius.) In the present work, essentially the same approach has been taken.

THE 906-I CHAMBER

The present work has been undertaken in part to verify the work of George and in general to follow the goal of precise, low-noise absolute measurements in the background level radiation fields. The design basis upon which we settled was a thin-walled, air-filled chamber with spherical geometry, as large as would be practical for field use. Such a design, as was already pointed out, avoids many of the difficulties of absolute calibration which may arise in more conventional ion chamber designs. In a thick-walled chamber there will be both attenuation of, and secondary production by, the incident radiation interacting with the wall material. The magnitude of such effects, as well as the ionization ratio of a chamber gas other than air, may or may not be accurately known.

On the other hand, thin walls preclude the use of high pressures. High gas pressures, and consequently thick walls, are conventionally used to enhance the ionization current relative to the unavoidable residual ionization which arises from traces of radioactive nuclides in the chamber walls. To minimize this contamination in a chamber operated near atmospheric pressure requires a large ratio of volume to surface area, hence a large volume and, preferably, a spherical geometry. The outer wall of the present chamber is an approximately 120-cm-diameter aluminum sphere, fabricated as two hemispherical spinings welded together around the equator.

The wall thickness varies, owing to cold flow in the spinning process, from 0.10 cm near the equator to 0.15 cm at the poles; the average value of 0.12 cm corresponds to 0.33 g/cm² of aluminum. A particle randomly incident on the chamber, therefore, traverses on the average a wall thickness of 0.66 g/cm². This thickness makes the chamber nearly opaque to beta radiation from terrestrial sources; the transmission of a beta

spectrum of 1.5-MeV endpoint energy will be of the order of 1%. For the cosmic radiation (and all but the very soft terrestrial gamma radiation), on the other hand, the wall is essentially transparent. One anticipates an attenuation of the cosmic ray soft component of less than 0.5%.

The collecting electrode is a 2.5-cm-diameter radial post, terminating in a 7.5-cm sphere concentric with the outer shell. Post and ball are both of aluminum and are electrically connected. The terminal assembly is shown in detail in Figure 1. It is more or less the conventional three-electrode configuration, the outermost flange, into which the central electrode mounts, acting as the shield or guard ring. The primary insulator is a Plexiglas ring cemented into a second aluminum flange, which in turn is welded to the chamber wall. The ground electrode extends into the chamber neck in such a way as to shield the active volume from the electric field on the main insulator. Gas inlets, etc., are provided in the fixed ground flange. In operation the whole assembly hangs at the lower pole of the spherical chamber. The ion current is measured between the central electrode and ground.

For absolute measurements of ionization density it is plainly necessary to know precisely the volume of the chamber. This cannot simply be calculated, as the spinning which forms the chamber's outer wall is not a perfect sphere. Two volume measurements were made. During fabrication the two hemispheres, not yet welded together, were braced, and their internal contour was carefully mapped; the resulting volume was calculated to be 904 ± 5 l. After completion of the chamber its volume was measured again by filling it with water and found to be 907 ± 2 l. (So as not to crush the chamber with the weight of the water, we carried out this measurement with the chamber floating in a swimming pool.) The weighted mean of the two measurements is 906.5 ± 2.2 l, corrected to 20°C.

The arrangement of electrodes at the chamber terminal is such that essentially all of this is active volume. The only inactive region will be that in the immediate vicinity of the chamber neck, where the electric field is shunted directly to the reentrant ground electrode; this is estimated to be less than a liter. Figure 2 shows a collecting voltage characteristic for the

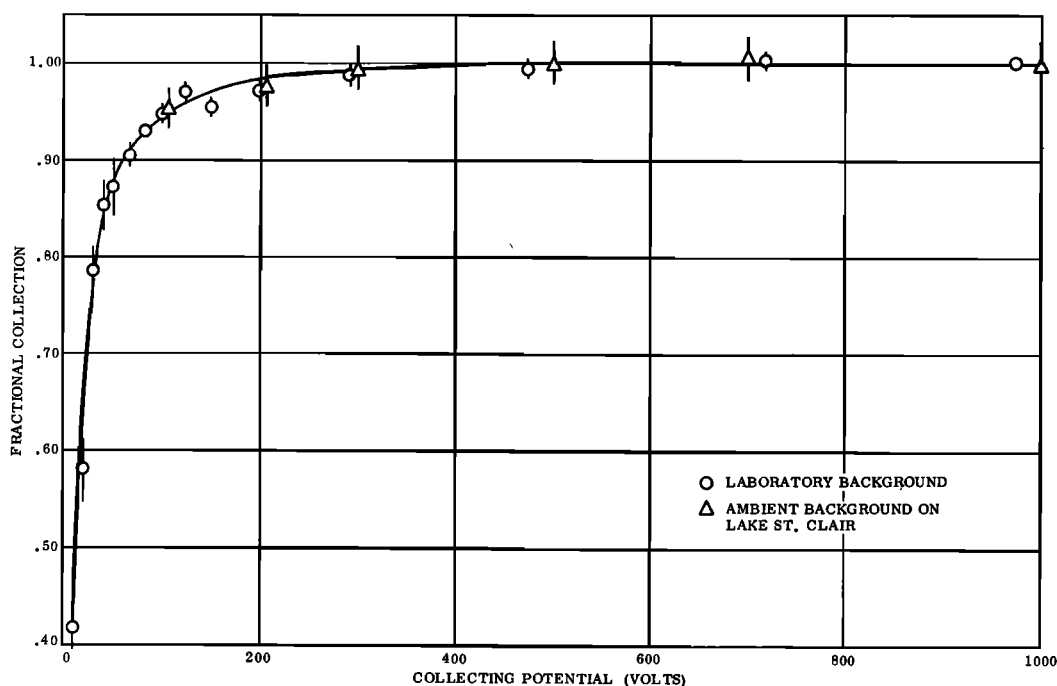


Fig. 2. Saturation characteristic of 906-l ionization chamber.

906-l chamber. It is quite flat above 400 V (about 0.43 V/cm at the chamber wall), confirming at least that no substantial volume lies in a region of marginal electric field.

In use, care must be taken to avoid contamination of the chamber reading by even slightly radioactive materials. Plastics used in the chamber and its mounting cradle, etc., contain no contemporaneous carbon, to avoid ^{14}C contamination. The chamber is filled from compressed air cylinders (cold trapped to remove oil and water vapor) which have been stored for at least several weeks and is operated at a slight overpressure to minimize inleakage. In fact, chamber leakage is negligibly small. When not in use, the chamber is kept sealed to avoid the deposition of atmospheric radon daughters. The contamination which is unavoidable is that due to radioactive trace elements in the wall material itself; this is discussed below.

MEASUREMENTS AND CALIBRATION

Measurements were carried out on freshwater lakes in order to minimize, as far as possible, effects due to the terrestrial gamma background. Mean free paths of 2.6-MeV photons (the 2.615-MeV line associated with the ^{232}Th series is the most energetic in the terrestrial background spectrum) are about 25 cm of water and 200 m of air. Consequently, sites were chosen in which data could be conveniently taken at least half a mile from shore, in a depth of at least 8 ft (2.4 m) of clear water. In September 1973 and in June and October 1974, runs were made on Anchor Bay of Lake St. Clair, about a mile offshore from Fair Haven, Michigan; and in June 1974, runs were made on Lake Huron, from 1 to 2 mi (1.6–3.2 km) east from Port Sanilac, Michigan. The water depth in Anchor Bay varied from 8 to 12 ft (2.4–3.6 m); that in Lake Huron was about 15 ft (4.5 m). In each case, in addition to the lake measurements, data were taken on a dock near shore. At Fair Haven this was about 35 ft (10.6 m) from shore, and at Port Sanilac it was about 200 ft (61 m). All data were taken in fair weather and in comparatively calm water.

The chamber was mounted in a Plexiglas cradle, which in turn was mounted to an aluminum baseplate approximately 1 cm thick. The arrangement was designed to minimize, as much as possible, the mass of scattering material near the ion chamber. In the 1973 run, chamber and cradle were mounted on a styrofoam raft; in the 1974 measurements a new fiberglass rowboat carried the chamber and auxiliary equipment. In each case the chamber was towed behind a cabin cruiser (the *Kel-lori*), with the experimenters alongside in a separate small boat. Usually, equipment and experimenters would be towed to a site, and data taken while adrift.

Determining the ionization density in an air-filled ion chamber requires, essentially, three simple measurements: the pressure in the chamber, its volume, and the current from it. In the present experiment, both chamber and ambient pressure measurements were made with Wallace and Tiernan differential manometers readable to a fraction of a torr, which were frequently cross-checked against one another and against station ambient pressure at the U.S. Weather Service station at Detroit Metropolitan Airport. The uncertainty of ± 1 torr which we have assumed for our pressure values is certainly conservative. In the June 1974 measurements a very small pressure leak in the chamber occurred; in those data we have assumed a somewhat larger uncertainty in chamber pressure.

The volume of the chamber is a slightly subtle question, as it is the active volume—the volume in which a saturation electric field exists when there is voltage on the chamber—that matters. However, the electrode configuration in the 906-l chamber is designed specifically to minimize inactive volume; as an indication of this, the saturation characteristic we observe is quite flat from 400 to 1000 V (see Figure 2). The difference between the physical volume of 906 ± 2 l and the active volume cannot be greater than a liter and is probably much less.

In ordinary background radiation fields the current from a chamber of this size, filled with near-STP air, will be a few tenths of a picoampere. In most of the measurements here

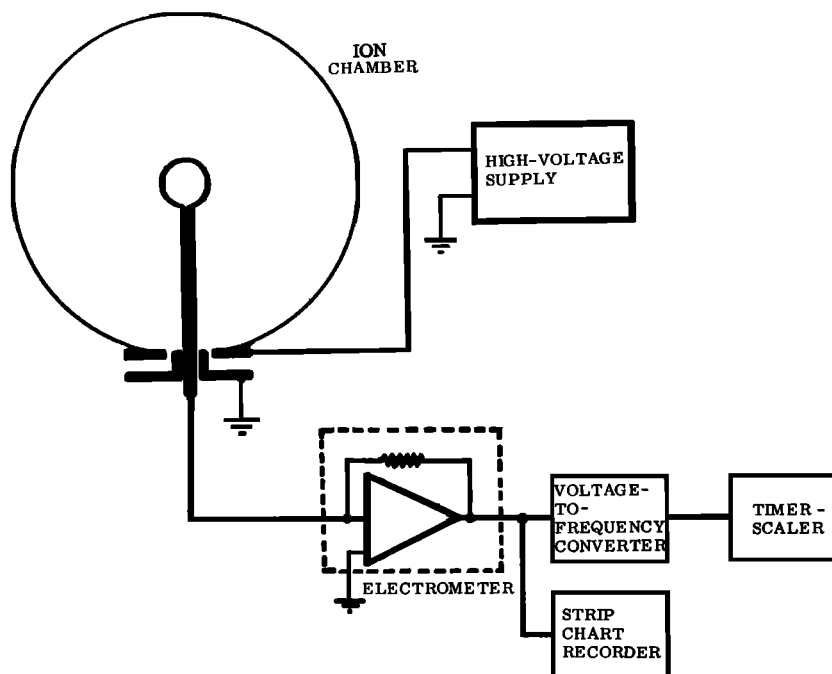


Fig. 3. Block diagram of measuring circuit.

reported, this current was measured by using a Victoreen model 475A vibrating capacitor input electrometer; the feedback resistor shown in Figure 3 was a Victoreen $5 \times 10^{11} \Omega$ (1%) resistor (the identical resistor to that used by *Shamos and Liboff* [1966]). For some of the June 1974 measurements a much faster MOSFET input solid state electrometer circuit employing a $10^{12} \Omega$ (2%) resistor was used. Our principal means of measuring the electrometer output voltage (of the order of several tenths of a volt) was to take 10-s counts of the output of a Dymec model 2210 voltage-to-frequency converter.

The chamber can operate in either polarity. To eliminate any spurious current due to an imbalance in the electrometer input circuit, we reversed polarity after every dozen or so readings; each reported value is a mean over several polarity reversals.

The electrometer circuits were calibrated by inserting a voltage source (Keithley model 260), which in turn was calibrated against a Leeds and Northrup type K3 potentiometer, into the electrometer feedback loop and measuring the output voltage with the voltage-to-frequency converter. The result was cross-checked against calibration directly at the electrometer input, by using a Keithley model 261 picoampere source, and against several auxiliary instruments. The calibration is self-consistent and, we believe, accurate, to within $\pm 1.5\%$.

The size of the present chamber is such that checking its calibration with a standard radioactive source was not considered feasible, as obtaining a satisfactory geometry would be quite difficult. We nevertheless believe that the systematic uncertainty in ionization density calibration due to pressure, volume, and current measurements cannot be greater than $\pm 2\%$, and we have included an uncertainty of this magnitude in the results reported below.

Because of the large volume of this chamber the fluctuations of the output current are quite small, even with air in the chamber. This very low noise level (the standard deviation of the fluctuations is of the order of 1% of the mean current level)

makes it possible to observe quite subtle effects. As an example, fairly large rapid excursions in ionization current have been observed with this device [*Kyker and Liboff*, 1974]. These may well result from cosmic ray air showers which are not ordinarily observable in ionization chambers close to sea level.

ENVIRONMENTAL GAMMA DOSIMETRY

We have carried out our measurements on freshwater lakes in order to minimize the terrestrial gamma ray component of the radiation background. However, an environmental gamma component is always present to some degree, if only from radon and thoron daughters in the atmosphere. We have more than once observed fluctuations in the chamber current, at times in the lake measurements, apparently correlated with variable offshore winds. We have attempted to measure this contribution to the chamber ionization density directly, by measuring environmental gamma ray spectra at the same location; in the October 1974 experiment the ionization chamber and gamma ray spectrometer measurements were carried out simultaneously, aboard the *Kellori*.

Figure 4 shows a typical spectrum of environmental gamma radiation, taken on land, measured by using a 4 in. $\times$ 4 in. NaI(Tl) scintillation detector, and a multichannel analyzer. The various contributors to the gamma background are all readily identifiable, as noted on the figure. The 2615-keV line (from ^{208}Tl) is characteristic of ^{232}Th daughters; lines at 610, 1120, 1764, and 2204 keV, all from the decay of ^{214}Bi , result from the presence of ^{238}U series nuclides, while ^{40}K produces a single line at 1460 keV. Many spectra also show lines from fission debris fallout, notably at 660 keV (^{137}Cs), 750 keV ($^{95}\text{Zr}/^{95}\text{Nb}$), and around 500 keV (from several different nuclides). Although the figure shows a spectrum on land, all the same component lines can usually be identified, with intensities an order of magnitude smaller, in the spectrum seen over water. The pattern of lines is sufficiently persistent and distinctive that the crystal spectrometer is self-calibrating.

Beck et al. [1966] have developed methods for inferring the

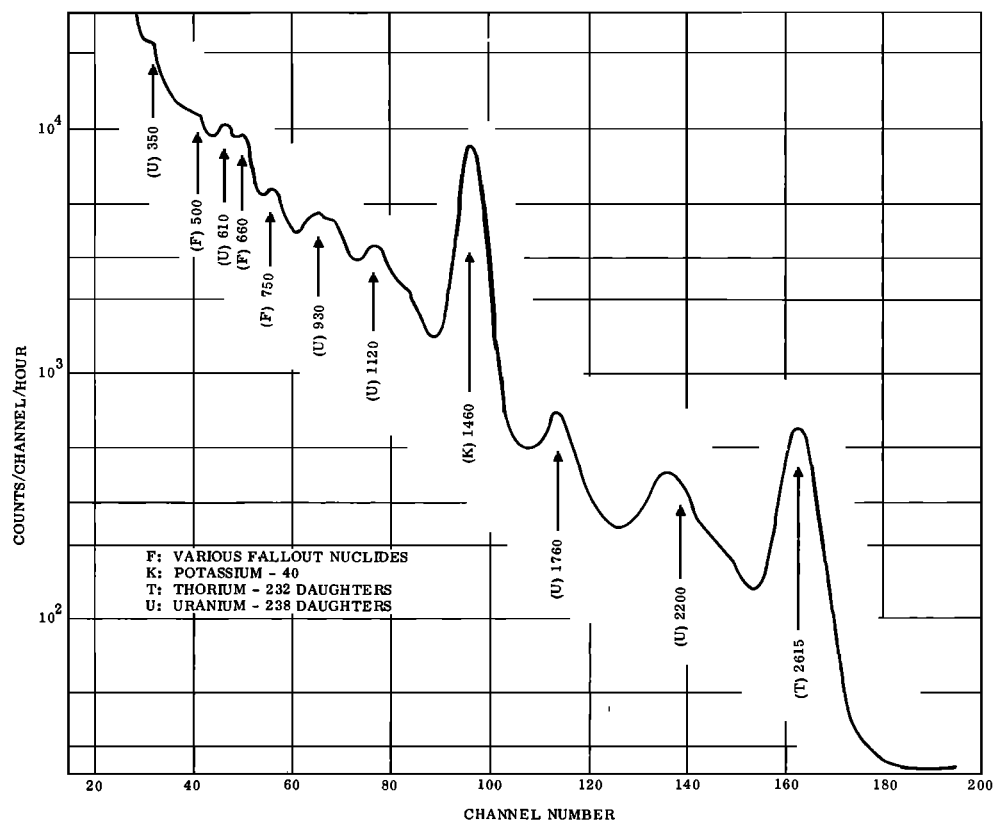


Fig. 4. Typical environmental gamma spectrum, taken with 4 in. $\times$ 4 in. NaI(Tl) scintillation spectrometer.

gamma dose rate directly from 4 in. $\times$ 4 in. NaI spectra and checked them against an extensive series of ionization chamber measurements. Their 'peak' method relates the gamma dose rate from each source to the total counts in the full-energy peak, above a continuum interpolated logarithmically under each peak; the 1764- and 2615-keV lines are used to measure the whole gamma dose from ^{238}U and ^{232}Th series nuclides, respectively. Their 'band' method infers dose rates from the total spectrum energy in specified bands about each of the landmark peaks. In analyzing spectra taken in the present work, both methods have been applied; in no case are the dose rates inferred by the two methods substantially different.

On several occasions we have carried out measurements of the terrestrial gamma dose at sites used for the ion chamber experiments. In all cases a Harshaw 4 in. $\times$ 4 in. NaI(Tl) crystal with integral photomultiplier was used in conjunction with a 1024-channel analyzer. The results, converted to ionization density, lay between 0.20 and 0.30 ion pairs/s cm^3 of air. Since all these measurements were made either on docks or on board the *Kellori* and since some ^{40}K and fallout contributions were always present, they cannot be considered inconsistent with the figure of 0.10 ion pairs/s cm^3 , due to radon and thoron daughters, observed by *Hess and Vancour* [1950] and calculated by *O'Brien* [1958]. Furthermore, the mean difference between lake and dock ion chamber results, at a given site, was 0.18 ± 0.08 ion pairs/s cm^3 in our experiments. We have consequently assumed the value of 0.10 ion pairs/s cm^3 in correcting those of our deep-water ion chamber measurements with which simultaneous gamma spectra were not taken; an average observed value of 0.25 ion pairs/s cm^3 is assumed for correcting the measurements made at docks.

The inference of gamma ray doses from NaI spectra de-

pends substantially on the spatial distribution of the source nuclides. *Beck et al.* [1966] have investigated this dependence for the case of fallout nuclides, where considerable variations in spatial distribution can be expected; they report as much as a factor of 2 variation between extreme cases. A good part of most of our measured gamma doses must be due to atmospheric radon and thoron, and substantial uncertainty in them might consequently be expected. On the other hand, the environmental gamma dose is a small contribution ($<15\%$) to the lake and mine measurements (described below), so a fairly large uncertainty is not damaging. As a consistency check, also, we have included in our results one measurement made on land, in which the gamma ray contribution is nearly half the total radiation background; the cosmic ray ionization inferred from that measurement is in excellent agreement with the lake measurements. Thus we are confident that the environmental gamma contribution to our measurements can be accurately accounted for.

CHAMBER BACKGROUND AND CONTAMINATION

Traces of naturally occurring radionuclides are present in essentially all materials; thus every radiation detector contributes, in some degree, to its own background. The effect of this self-contamination on the 906-I chamber is substantial. We have attempted to measure this contamination both directly—in a very low background environment—and indirectly, by an analysis of the noise in the chamber signal.

The direct measurement was carried out with the cooperation of the Morton Salt Company in their Painesville, Ohio, salt mine at a depth underground of approximately 600 m. At this depth the cosmic ray contribution to the natural background is, to all intents and purposes, completely shielded out.

TABLE 2. Emission Properties of Wall Contaminations

Source Case	$k_\alpha = I/n_\alpha$, ion pairs/ α	n_α/μ , cm ³	I/μ , ion pairs cm ³	$k_\beta = I/n_\beta$, ion pairs/ β	n_β/μ , cm ³	I/μ , ion pairs cm ³	I_β/I_α
Single α decay, $E_\alpha = 5.5$ MeV	9.0×10^4	56	5.0×10^6				
Single β decay, $E_\beta = 600$ keV				5.5×10^3	420	2.3×10^6	
²³⁸ U (old) and all daughters in equilibrium	9.3×10^4	440	4.1×10^7	5.9×10^3	3600	2.1×10^7	0.51
²³⁸ U (young) and two daughters in equilibrium	6.7×10^4	38	2.5×10^6	4.9×10^3	330	1.6×10^6	0.64
²²⁶ Ra and all daughters in equilibrium	10.0×10^4	310	3.1×10^7	6.0×10^3	3200	1.9×10^7	0.61
²³² Th and all daughters in equilibrium	10.7×10^4	390	4.1×10^7	6.1×10^3	3100	1.9×10^7	0.46

The ionization in the chamber thus consists just of the environmental gamma radiation and the chamber's own self-contamination. As the surroundings were essentially pure halite for several feet in every direction, the environmental gamma component is expected to be small; it was measured simultaneously, using the NaI spectrometer, as 0.13 ± 0.02 ion pairs/s cm³. A chemical assay revealed that the potassium concentration in samples of the halite from this mine was 1.1×10^{-4} g(K)/g(NaCl). The resulting estimate of the ⁴⁰K contribution to the gamma background was of the same order of magnitude as that observed. Radon and thoron daughters were also observed in the mine spectra, owing, we believe, to the fact that the equipment was located near the main ventilator intake for the entire mine. The amounts were of an order comparable to those observed over lakes. The mean of several measurements of the total ion chamber current was 1.12 ± 0.05 ion pairs/s cm³, so we find directly for the residual chamber current, due to self-contamination, a value of 0.99 ± 0.05 ion pairs/s cm³.

In low-level ionization chamber measurements the bulk of the residual ionization is usually considered to arise from traces of alpha-emitting nuclides in the chamber wall material. This alpha activity, however, almost certainly is due to contaminant nuclides in the natural uranium and thorium series. This being the case, there will also be a contribution due to beta particles emitted by other series nuclides. Since their mean free path is far longer, the contribution of the betas may be important only in a large chamber and consequently is sometimes overlooked. The alpha activity can be estimated from the chamber signal-to-noise ratio, but since each event produces far fewer ions, betas add little to the fluctuations of the chamber current.

Because this contribution cannot be measured directly, we have attempted to calculate it. Given the source nuclides and the chamber geometry, the ion current produced is calculable. In a spherical chamber, from a contaminant uniformly distributed in the wall material, we may write (for either an alpha or a beta contamination)

$$I = \frac{4\pi R^2 \mu}{\eta} \int_0^{E_0} dE_2 \int_{E_2}^{E_0} dE_1 \int_0^{\pi/2} d\theta \frac{f(E_1, E_0)}{\epsilon(E_2)} E_c(E_2, \theta) \sin \theta \cos \theta \quad (1)$$

while the number of particles per second emitted into the chamber is

$$n = \frac{I}{k} = 2\pi R^2 \mu \int_0^{E_0} dE_2 \int_0^{E_0} dE_1 \frac{f(E_1, E_0)}{\epsilon(E_2)} \quad (2)$$

In both, k is the average number of ion pairs per particle, R is the chamber radius, η is the average energy per ion pair in the chamber gas, μ is the activity density (in decays/s/cm³) in the wall material, E_0 is the particle energy (endpoint energy for betas), E_1 is the energy of decay (equal to E_0 for alphas), E_2 is the particle energy at wall gas boundary, $\epsilon(E)$ is the specific energy loss (in MeV/cm) in the wall material at energy E , $f(E, E_0)$ is the distribution of decay energies, and $E_c(E, \theta)$ is the energy delivered to chamber gas by a particle surfacing with energy E and angle θ to the wall normal.

For alpha particles, $f(E, E_0)$ can be approximated by a delta function, and as most alphas will be stopped in a few centimeters of air, we may put $E_c = E_2$. Most betas, however, will not be stopped in the chamber, and $E_c(E_2, \theta)$ is dominated by geometry; in addition, a realistic decay energy distribution must be used.

In the present case, of course, we have no knowledge a priori of the specific nature of the contamination or of the density μ . Results of (1) and (2), integrated numerically, are shown relative to μ in Table 2 for several plausible source cases. For sources, such as the natural decay series, yielding both alphas and betas the ratio of beta- and alpha-induced residual ion currents is of course independent of μ . For most of the plausible contaminations in the natural series this ratio falls between 0.45 and 0.70. Given some separate means of inferring the alpha-induced part, the total residual ionization is thus approximately predictable.

Such a means is provided by an analysis of the fluctuations of the chamber signal. Following *Shamos and Liboff* [1968], we write the residual ionization in the form

$$I = n_\alpha k_\alpha + n_\beta k_\beta \quad (3)$$

where for each particle, n is the number per second introduced into the chamber and k the average number of ion pairs per particle. If the time constant of the measuring instrumentation is τ , the standard deviation of a measurement of the current will be

$$\sigma_I = [(n_\alpha k_\alpha^2 + n_\beta k_\beta^2)/2\tau]^{1/2} \quad (4)$$

A minor complication arises because only a fraction of the alpha-induced ionization is collected, owing to columnar re-

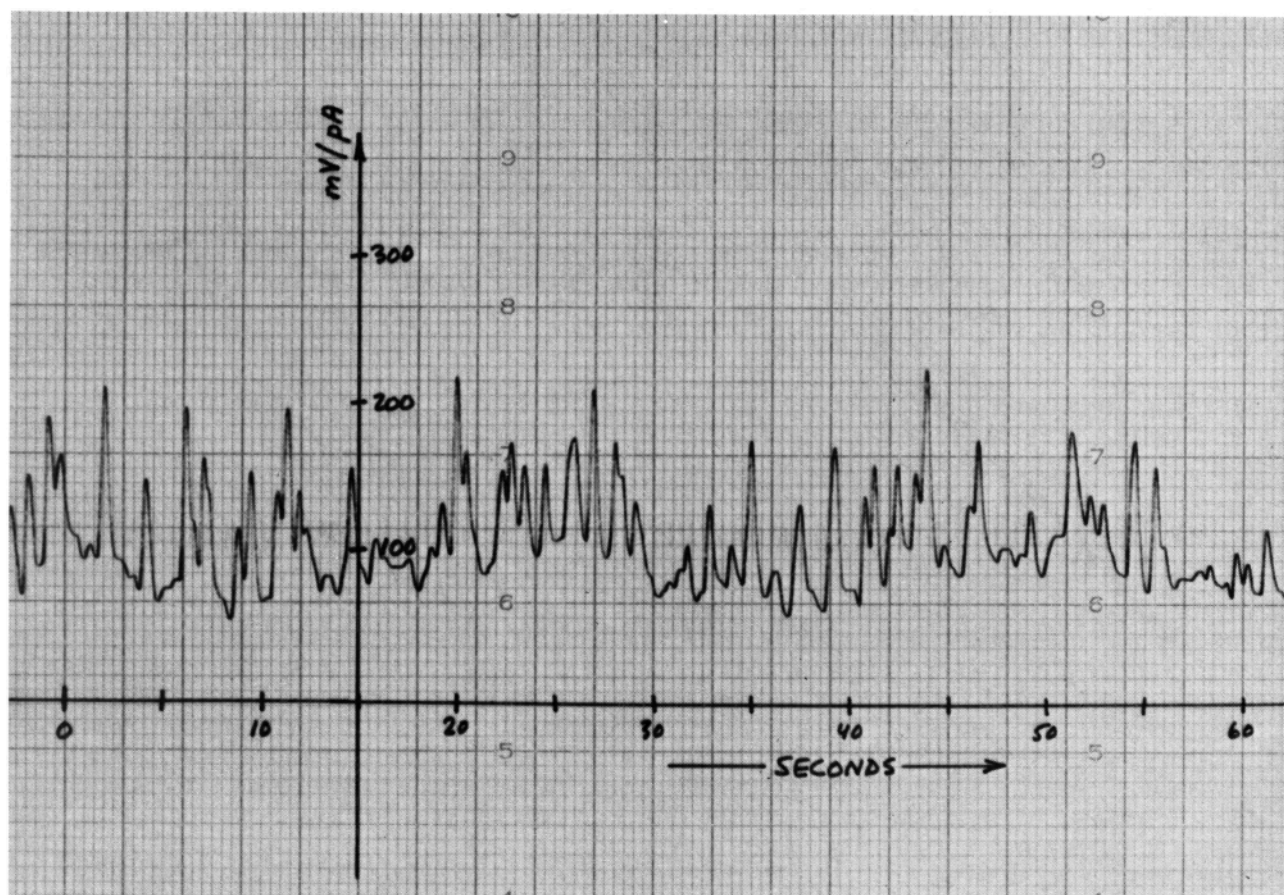


Fig. 5. Ion current record in Painesville salt mine; taken with fast ($\tau \sim 0.3$ s) MOSFET electrometer to enhance noise.

combination at the chamber walls. The effect is to reduce k_α . As we usually operate near 1-V/cm field at the chamber wall, we expect the fraction collected to be around 0.53 (Jaffe [1913, 1929a, b], reviewed by Boag [1956]). From the calculations presented in Table 2 we take as representative values $k_\alpha = 9.1 \times 10^4$, $k_\beta = 5.8 \times 10^8$, and $I_\beta/I_\alpha = (n_\beta k_\beta)/(n_\alpha k_\alpha) = 0.58$. Using these values and eliminating n_α between (3) and (4), we may simply relate I , σ_I , and τ .

During the mine experiment the standard deviation of the chamber signal was measured at a variety of collecting voltages, using a fast ($\tau \sim 0.3$ s) MOSFET input electrometer circuit to enhance the noise. A sample of this data is shown in Figure 5. Near our usual range of operating voltages (700–900 V) we find a value of $\sigma_I = (3.16 \pm 0.15) \times 10^6$ ion pairs/s. Allowing a reasonable range of uncertainty for the uncertain contaminant nuclides, the above procedure predicts a chamber current of $(0.96 \pm 0.25) \times 10^6$ ion pairs/s or 1.06 ± 0.27 ion pairs/s cm^3 in the 906-l chamber. This is in excellent agreement with the measured value of 0.99 ± 0.05 ion pairs/s cm^3 for the residual ionization. (This analysis, of course, neglects the possibility of contaminants which emit only betas.)

Accepting the measured result, the implied alpha activity of the aluminum wall is $0.69 \pm 0.20 \alpha/\text{cm}^2 \text{ h}$, or $8.7 \pm 2.5 \alpha/\text{s}$ in the chamber. Although large, this number is not wholly inconsistent with other evidence. In Figure 5, something like 1.5 or 2 peaks, corresponding to individual alpha events, per second can be seen, and it is certain that many of the smaller events will be unresolved. Published values for the alpha activity of commercial aluminum are highly variable. A 1961 review [Devoe, 1961] listed values ranging from 0.07 to 0.79 $\alpha/\text{cm}^2 \text{ h}$,

while another source [Sharpe, 1964] quotes values of 0.31 and 0.2; the median of reported values is about 0.2 $\alpha/\text{cm}^2 \text{ h}$. Also, in experiments conducted in the summer of 1974 with 13-l cylindrical chambers we observed a systematic difference between readings of identical aluminum and plastic chambers. The wall alpha activity of the aluminum required to explain the observed difference was $0.35 \pm 0.15 \alpha/\text{cm}^2 \text{ h}$.

Although the residual ion current due to wall contamination in the 906-l chamber is large, we believe that it has been measured accurately enough so as not to affect measurements made with this device.

RESULTS

Table 3 presents a summary of the results of all our measurements and the corrections made to the raw ion current observed. All currents are given in units of ion pairs per second per cubic centimeter of STP air. I_{obs} is the observed ionization density; each value represents the mean of several hundred 10-s samples of the chamber current. The errors shown include both the statistical error of these readings and the uncertainties in chamber calibration. I_γ is the estimated contribution to I_{obs} of environmental gamma radiation. Through June 1974, gamma ray spectra were not taken simultaneously with ion chamber measurements, and I_γ is inferred from subsequent measurements on the same sites. In the last three cases, I_γ is taken from simultaneous gamma spectra. I_{ch} is the contribution to I_{obs} due to the residual ionization of the chamber itself, as discussed above; it varies very slightly because the chamber's sensitivity to the beta-induced component depends on chamber pressure.

TABLE 3. Summary of All Measurements

Experiment	I_{obs}	Corrections		I	Sea Level STP I_0
		I_γ	I_{ch}		
Sept. 6-7, 1973, Lake St. Clair (lake)	2.91 ± 0.08	(0.10 ± 0.06)	0.98 ± 0.06	1.83 ± 0.11	1.84 ± 0.11
Sept. 7, 1973, Lake St. Clair (dock)	3.18 ± 0.09	(0.25 ± 0.12)	0.98 ± 0.06	1.95 ± 0.14	1.97 ± 0.14
June 27, 1974, Lake St. Clair (lake)	3.21 ± 0.11	(0.10 ± 0.06)	0.96 ± 0.05	2.15 ± 0.13	2.29 ± 0.14
June 27, 1974, Lake St. Clair (dock)	3.28 ± 0.12	(0.25 ± 0.12)	0.96 ± 0.05	2.07 ± 0.16	2.20 ± 0.17
June 28, 1974, Lake Huron (lake)	3.03 ± 0.13	(0.10 ± 0.06)	0.96 ± 0.05	1.97 ± 0.14	2.10 ± 0.15
June 28, 1974, Lake Huron (dock)	3.32 ± 0.13	(0.25 ± 0.12)	0.96 ± 0.05	2.11 ± 0.17	2.25 ± 0.17
July 31, 1974, Rochester, Michigan (land)	5.81 ± 0.20	2.75 ± 0.12	0.96 ± 0.05	2.10 ± 0.24	2.15 ± 0.24
October 4, 1974, Lake St. Clair (lake)	3.48 ± 0.11	0.21 ± 0.02	1.00 ± 0.05	2.27 ± 0.13	2.29 ± 0.13
October 4, 1974, Lake St. Clair (dock)	3.59 ± 0.11	0.29 ± 0.03	1.00 ± 0.05	2.28 ± 0.13	2.30 ± 0.13

The average of all the values in the last column is 2.15 ± 0.05 .

The measurements of I_a in parentheses were not made simultaneously with I_{obs} .

When these contributions are subtracted, the result I is the ionization intensity due to the cosmic radiation. Finally, I is corrected to I_0 , the sea level cosmic ray ionization in STP air, in the last column. In making the correction to sea level we have assumed a barometer coefficient of 0.40% per torr. This value is supported by the measurements both of *Shamos and Liboff* [1966] and of *George* [1970], with respectively thicker- and thinner-walled chambers than the present work. The mean and standard error of the nine values shown for I_0 is $I_0 = 2.15 \pm 0.05$ ion pairs/s cm^2 , and we report this value for the sea level ionization intensity in standard air. This is in excellent agreement with the recent values in Table 1, except for the value of *George*, and with the value of 2.22 calculated by *O'Brien* [1970]. Finally, as it bears on the stated accuracy of our result, we note that the standard deviation of the nine I_0

values in Table 3 is 0.14, which is completely consistent with the estimated standard errors of the individual determinations.

DISCUSSION

It is interesting to examine carefully the set of available readings on the cosmic ray sea level ionization. Of particular concern is the extent to which modulation and weather effects may have influenced these readings. For example, *Bhattacharyya* [1974] has shown that the muon spectrum and intensity vary appreciably with solar activity. In Figure 6 we have plotted, from Table 1, those values obtained after 1960, along with the solar modulation for the same period, as evidenced by the mean annual levels of the Climax neutron monitor [*Fulks*, 1975]. Additional points include the value that we report in this paper, 2.15 ion pairs/s cm^2 , as well as the

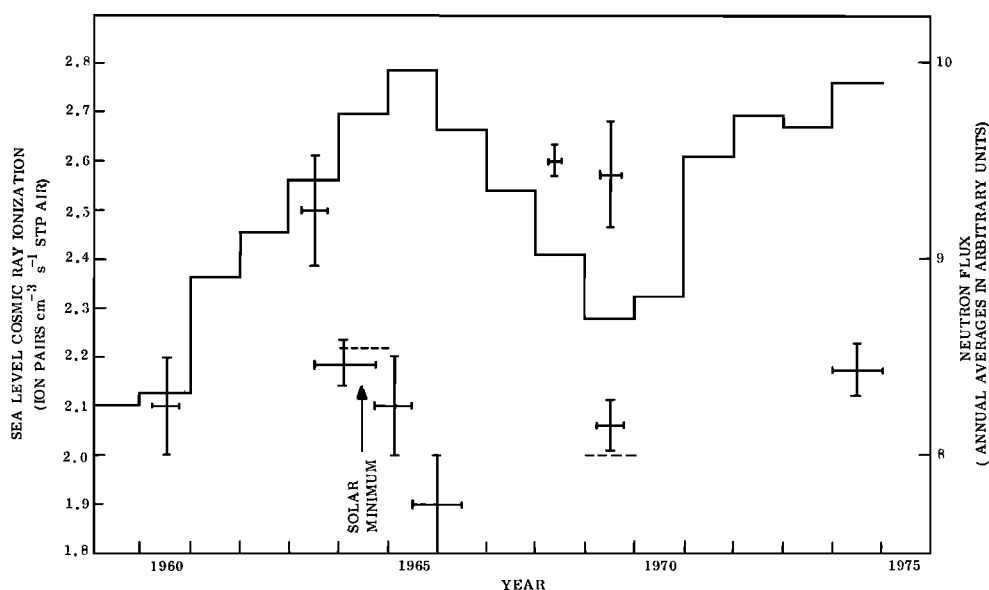


Fig. 6. Values of sea level cosmic ray ionization after 1960. Horizontal bars indicate actual periods of measurement; vertical bars error estimates. The two dashed lines represent theoretical estimates by *O'Brien* [1975] for solar minimum and maximum. The solar modulation for the period is also shown, indicating that the wide scatter in ionization is independent of the modulation.

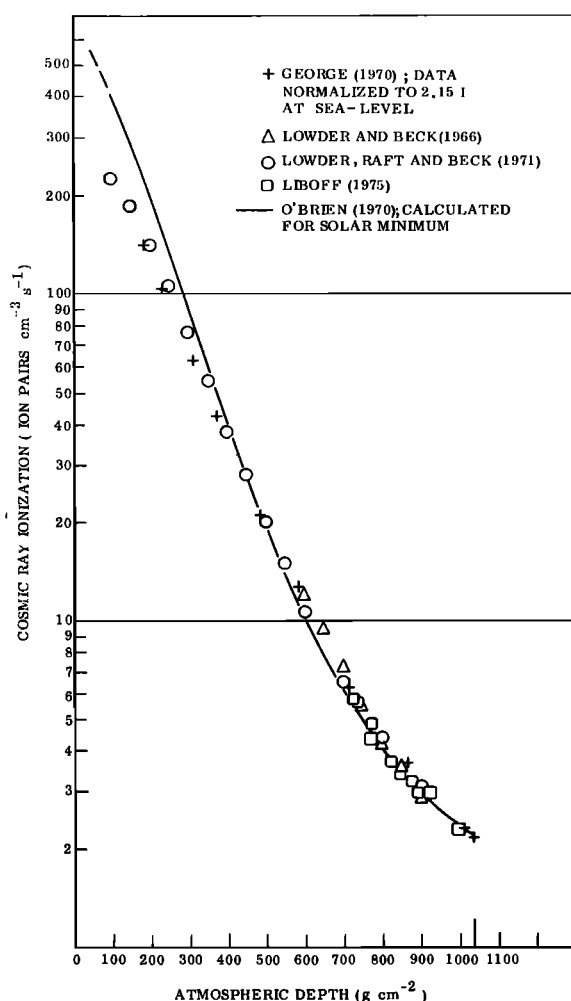


Fig. 7. Altitude variation of cosmic ray ionization, after normalizing data by George [1970] to 2.15 ion pairs/s cm^{-3} at sea level. The lack of agreement at higher altitudes reflect the fact that the measurements [George, 1970; Lowder et al., 1971] were obtained during solar maximum, whereas the calculation by O'Brien [1970] applies to the solar minimum.

theoretical values obtained by O'Brien [1975] for solar maximum and solar minimum, 2.00 and 2.22 ion pairs/s cm^{-3} , respectively. The horizontal bars indicate the actual period of measurement, and the vertical bars represent error estimates.

It is quite clear that solar modulation cannot be regarded as the cause of the scatter. There seems to be, instead, a roughly bimodal distribution of values. For the lower set, all of the values save one are found within the bounds predicted by O'Brien. In particular, it is possible to specify four estimates for the sea level cosmic ray ionization during solar minimum, independently arrived at, using different techniques, and all included within each other's error bars: 2.18 ± 0.05 ion pairs/s cm^{-3} [Shamos and Liboff, 1966], 2.1 ± 0.1 ion pairs/s cm^{-3} [Lowder and Beck, 1966], 2.22 ion pairs/s cm^{-3} [O'Brien, 1970], and 2.15 ± 0.05 ion pairs/s cm^{-3} (present work).

The discrepancy between our result and that of George [1970], the only other directly comparable measurement in air, is difficult to pinpoint. On the basis of the design and characteristics of his ionization chamber it is unlikely that there was appreciable internal radioactive contamination. The data presented in Table 3 tend to indicate that wooden docks have a small but measurable gamma activity compared to open wa-

ters nearby. This difference, of the order of 0.2 ion pairs/s cm^{-3} may be due to fallout retention or perhaps intrinsic ^{40}K activity. The George value, in large measure, is derived from measurements on a dock.

It is interesting to realize that the difference between the published George value, 2.60 ion pairs/s cm^{-3} STP air, and that reported here, 2.15 ion pairs/s cm^{-3} STP air, is about 17%, which is coincidentally the estimated percentage calibration error that Carmichael [1971] suggested for the California Institute of Technology ionization chambers in the period since 1951. It is instructive to attempt to renormalize George's altitude data to our sea level value of 2.15 ion pairs/s cm^{-3} . We present this renormalized altitude variation in Figure 7, along with a number of measurements by other observers.

The most vexatious part of the present work was that a wall material was chosen for the ionization chamber which, relatively speaking, was rather contaminated. However, in view of the excellent signal-to-noise ratios which we observed, further use of this device is warranted, especially for precise measurements on the low-level radiation background. It is hoped that we may use an alternate wall material to aluminum, capable of being formed in large dimensions and lower in intrinsic radioactivity.

Acknowledgments. The authors are very grateful for the capable assistance of several research assistants, among them M. Karas, E. Chaffee, D. Kaufeld, M. Williams, and G. Persha. We would also like to thank, for his generous assistance during the mine experiment, the chief engineer of the Morton Salt Co., B. Cummings. We are indebted to K. Waatt and S. Miller of the Department of Chemistry at Oakland University, who analyzed halite specimens from the Morton mine for potassium content. The skills of V. Wendler and F. Kopetko of the Oakland University Machine Shop in fabrication of the ion chamber and the cooperation of F. Kopetko (owner of the Kellori) throughout the measurements are gratefully acknowledged. We also thank W. M. Lowder for several helpful conversations.

REFERENCES

- Beck, H. L., W. M. Lowder, B. G. Bennett, and W. J. Condon, Further studies of external environmental radiation, *Rep. HASL-170*, U.S. At. Energy Comm. Health and Safety Lab., New York, March 1966.
- Bhattacharyya, D. P., Influence of long term solar modulation on the relativistic muon intensity, *J. Phys. A*, 7, 2333-2337, 1974.
- Boag, J. W., Ionization chambers, in *Radiation Dosimetry*, edited by G. J. Hine and G. L. Brownell, Academic, New York, 1956.
- Carmichael, H., Comments on the absolute ionization of cosmic radiation at balloon altitudes covering four solar maxima, paper presented at 12th International Conference on Cosmic Rays, Hobart, Tasmania, Australia, 1971.
- Devoe, J. R., Radioactive contamination of materials used in scientific research, *Rep. 34*, Nat. Acad. of Sci., Nat. Res. Council, Washington, D. C., 1961.
- Fulks, G. J., Solar modulation of galactic cosmic ray electrons, protons and alphas, *J. Geophys. Res.*, 80, 1701-1714, 1975.
- George, M. J., New data on the absolute cosmic ray ionization in the lower atmosphere, *J. Geophys. Res.*, 75, 3693-3705, 1970.
- Gustafson, P. F., J. Kastner, and J. Lützelshwab, Environmental radiation: Measurement of dose rates, *Science*, 145, 44-47, 1964.
- Herbst, W., Investigations of environmental radiation and its variability, in *The Natural Radiation Environment*, edited by J. A. S. Adams and W. M. Lowder, University of Chicago Press, Chicago, 1964.
- Herbst, W., and G. Hübner, Untersuchungen über die durchdringende äusseren Umgebungsstrahlung, *Atomkernenergie*, 6, 75-81, 1961.
- Hess, V. F., and R. P. Vancour, The ionization balance of the atmosphere, *J. Atmos. Terr. Phys.*, 1, 13-25, 1950.
- Jaffe, G., Zur Theorie der Ionisation in Kolonnen, *Ann. Phys. Paris*, 42, 303-344, 1913.
- Jaffe, G., Kolonnenionisation in Gasen bei erhöhtem Druck, *Phys. Z.*, 30, 849-856, 1929a.
- Jaffe, G., Zur Theorie der Ionisation in Kolonnen, II, *Ann. Phys. Paris*, Ser. 5, 1, 977-1008, 1929b.

- Kyker, G. C., and A. R. Liboff, Thin-walled ionization chamber measurements of cosmic and environmental radiation, Second Workshop on the Natural Radiation Environment, *Rep. 287*, U.S. At. Energy Comm. Health and Safety Lab., New York, 1974.
- Liboff, A. R., Cosmic ray ionization in the lower atmosphere, in *The Natural Radiation Environment II, Conf.-720805-P1 and P2*, edited by J. A. S. Adams, W. M. Lowder, and T. F. Gesell, Energy Research and Development Administration, Germantown, Md., 1975.
- Lowder, W. M., and H. L. Beck, Cosmic ray ionization in the lower atmosphere, *J. Geophys. Res.*, **71**, 4661-4668, 1966.
- Lowder, W. M., P. D. Raft, and H. L. Beck, Experimental determination of cosmic ray charged particle intensity profiles in the atmosphere, paper presented at Symposium on Natural and Man-Made Radiation in Space, Las Vegas, Nev., March 1971.
- Millikan, R. A., Cosmic ray ionization and electroscope constants as a function of pressure, *Phys. Rev.*, **39**, 668-669, 1932.
- Millikan, R. A., and G. H. Cameron, A more accurate and extended cosmic ray ionization-depth curve, and the present evidence for atom-building, *Phys. Rev.*, **37**, 235-252, 1931.
- Millikan, R. A., and H. V. Neher, World survey of sea-level cosmic ray intensities, *Phys. Rev.*, **50**, 15-24, 1936.
- O'Brien, K., Some variable contributors to natural background, *Rep. 27*, U.S. At. Energy Comm. Health and Safety Lab., New York, 1958.
- O'Brien, K., Calculated cosmic ray ionization in the lower atmosphere, *J. Geophys. Res.*, **75**, 4357-4359, 1970.
- O'Brien, K., The cosmic ray field at ground level, in *The Natural Radiation Environment II, Conf.-720805 P1*, edited by J. A. S. Adams, W. M. Lowder, and T. F. Gesell, Energy Research and Development Administration, Germantown, Md., 1975.
- O'Brien, K., and J. R. McLaughlin, The radiation dose to man from galactic cosmic rays, *Health Phys.*, **22**, 225-232, 1972.
- Ohlsen, H., Bestimmung der mittleren Bevölkerungsbelastung durch natürliche äussere Strahlung auf dem Gebiet der DDR, *Rep. SZS-14/69*, Staatliche Zent. für Strahlenschutz, Berlin, 1969.
- Neher, H. V., Recent data on geomagnetic effects, in *Progress in Cosmic Ray Physics*, pp. 243-314, Interscience, New York, 1952.
- Raft, P. D., W. M. Lowder, and H. L. Beck, Measurements of cosmic ray ionization in the atmosphere, 1968-1970, *Rep. HASL-234*, U.S. At. Energy Comm. Health and Safety Lab., New York, Aug. 1970.
- Shamos, M. H., and A. R. Liboff, A new measurement of the intensity of cosmic ray ionization at sea level, *J. Geophys. Res.*, **71**, 4651-4659, 1966.
- Shamos, M. H., and A. R. Liboff, New ionization chamber technique for the measurement of environmental radiation, *Rev. Sci. Instrum.*, **39**, 223-229, 1968.
- Sharpe, J., *Nuclear Radiation Detectors*, 2nd ed., p. 105, John Wiley, New York, 1964.
- Solon, L. R., W. M. Lowder, A. Shamos, and H. Blatz, Investigations of natural environmental radiation, *Science*, **131**, 903-906, 1960.
- Yeates, D. B., A. S. Goldin, and D. W. Moeller, Natural radiation in the urban environment, *Nucl. Safety*, **13**, 275-286, 1972.

(Received September 2, 1975;
revised July 3, 1978;
accepted July 24, 1978.)